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X-Ray Emission Produced in Charge-Exchange Collisions between Highly Charged Ions and Argon: Role of the Multiple Electron Capture

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X-ray emission produced in charge-exchange collisions between highly charged ions and argon: Role of the multiple electron capture

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In this work we use the classical trajectory Monte Carlo method within an eight-electron scheme to theoretically study photonic spectra that follow charge-exchange processes between highly charged ions of charge states 10+, 17+, 18+, and 36+ with neutral argon. The energy range considered is 18 eV/amu to 4 keV/amu, covering typical electron beam ion traps and solar wind energies. The role played by multiple electron capture processes for the different collision systems under consideration is explicitly analyzed and its contribution separated as arising from radiative decay and autoionizing multiple capture. For the present collision systems we find that multiple electron capture is responsible for 50%–60% of the resulting x-ray spectra. The present results are of direct relevance to the astrophysical program.

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I. INTRODUCTION

The upcoming launch of the Astro-H mission, scheduled for 2015, will provide astrophysicists with high-resolution x-ray spectra (<7 eV resolution in the 0.3–12 keV band) of different deep sky objects. These observations are expected to improve our knowledge on the structure and evolution of the Universe [1]. This ultimate and highly sophisticated mission has transited from its very beginning (initial design and expected performance), the road paved by previous orbiters such as ROSAT, XMM-Newton, and the Chandra X-ray Observatory. The low-resolution x-ray spectrometer of ROSAT (300 eV resolution) led us to the unexpected detection of x-ray emission from comet P/Hyakutake in 1996 [2]. A new generation of orbiters—Chandra and XMM-Newton, with x-ray charge-coupled device (CCD) detectors that provided a much improved resolution in the order of 100 eV—was needed before the origin of the x-ray emission in comets could be fully understood. By the year 2001, Chandra provided clear evidence on the dominant role of the charge-exchange mechanism followed by photonic emission proposed by Cravens in 1997 [3]. The spectrum of comet C/LINEAR 1999 S4 revealed emission lines close to those predicted for charge exchange between fully stripped and hydrogenic C, N, and O ions with neutral targets in the coma [4]. Since then, these orbiters surveyed several cases of cometary and planetary x rays [5].

By 2005, an x-ray microcalorimeter spectrometer with high-energy resolution (<7 eV) was launched in the Suzaku mission. It would have provided high-resolution spectra had not it been that the cryogenic system broke down within the first two weeks, degrading the performance of the x-ray spectrometer. Ten years would go by before another mission would be ready to tackle these objectives.

Since the very beginning of this century laboratories worldwide have been working on the study of charge-exchange collision processes with the goal to recreate cometary spectra based on laboratory data [6–9]. Electron beam ion traps (EBIT traps) were married at Lawrence Livermore National Laboratory (LLNL) to the spare calorimeter spectrometer of the Suzaku mission which was calibrated at Goddard Space Center. This setup provided *in situ* spectra with an unprecedented resolution of about 8 eV at collision energies which are two orders of magnitude below those corresponding to the solar wind. Linear accelerators, on the other hand, were used at the Jet Propulsion Laboratory and reached solar wind velocities but missed the dominant forbidden lines $2\,^3S \rightarrow 1\,^1S$ which are long lived and cannot take place within the typical dimensions of the devices under use. Since then, a limited but nonetheless important set of data has been collected by several experimental groups.

From the astrophysical side, the necessity of accurate line emission cross sections relies on the fact that solar wind ion abundances can be estimated by deconvoluting the measured spectra. By doing so, a solar wind map can be performed for regions that cannot be reached by orbiters. In cometary x-ray spectra, the solar wind ion abundances are considered as fitting parameters, and estimations for the solar wind ion abundances can be considered reliable only if accurate line emission cross sections are used [10,11].

Theoretical studies can provide, in principle, much assistance to this task. Studies have been mainly conducted by means of the classical overbarrier (COB) model [7], the Landau-Zener multichannel (LZMC) model [12], and the classical trajectory Monte Carlo (CTMC) method [13–18]. In addition, electron capture cross sections for hydrogen targets obtained by means of atomic orbital close coupling (AOCC) codes have been used by Bodewits and Hoekstra to simulate cometary spectra [9] and line emission studies have been also recently made within the two-center–basis generator model (TC-BGM) [19]. However, the COB, LZMC, and TC-BGM models need to use *ad hoc l* distributions, like the flat and the statistical distributions, for the projectile population. In contrast, CTMC inherently keeps track of the electronic momenta throughout the collision process.

Cometary spectra dominated by N, C, and O lines have been successfully reproduced in previous works by taking into account the solar wind ion abundances provided by the

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Advanced Composition Explorer, the CXO ACIS-S spectrometer effective area, and the corresponding line emission cross sections provided by the three-body CTMC method [13,14]. However, a significant problem appears for highly charged solar wind ions colliding with multielectronic targets which is the effect of multiple electron capture. As multiple capture probabilities increase with the projectile charge, a key point during the analysis of laboratory and astrophysical spectra is the accurate determination of the fraction of x rays that originate in multiple electron transfer [16].

In this work, we extend our previous work and explicitly evaluate the role of the multiple electron capture process in the photonic spectra which follow charge-exchange collisions involving highly charged ions and argon targets. This target has been explored at LLNL [20], Berlin-EBIT [21], and National Institute of Standards and Technology (NIST) [22], hence providing a good benchmark for theoretical analysis. It is expected that success in the proper description of these laboratory spectra will provide useful information for future studies of more complex cometary neutrals like H_2O and CO_2. In Sec. II we describe the CTMC model and the scheme used to account for the Auger cascading processes which ultimately lead to autoionizing multiple electron capture and radiative decay. In Sec. III we show our main results. Conclusions and outlook are drawn in Sec. IV.

II. THEORETICAL METHOD

Theoretical electron capture and line emission cross sections have been calculated using the CTMC method [23,16]. Hamilton's equations are solved for a system composed by the projectile, the ionic core, and eight noninteracting electrons which are sorted according to the sequential binding energies for the target. This procedure models the sequential electron removal of electrons up to the whole M shell. Although electron-electron correlations are not accounted in detail, the energy deposition needed for many-electron removal is properly considered. Electrons are sorted according to their quantum mechanical momentum distribution and their interaction with the ionic core is considered to be Coulomb-like with an effective charge that is set in order to provide the best possible agreement with the quantum mechanical radial distribution. In a previous work, for which only three electrons were explicitly considered, we have shown that the initialization of events over the correct quantum mechanical momentum distribution correctly leads to the nodal structure in the radial distribution [16].

For the sake of simplicity, hydrogenic energy levels are used to determine the projectile population after multiple charge exchange.

The Auger decay scheme used in this work is based in the decay scheme developed by Ali *et al.* [24] for collisions involving argon ions with argon atoms and can be summarized as follows:

(i) Multiply excited states dominantly stabilize via multiple Auger processes.

(ii) Two-electron Auger processes are considered only.

(iii) Transitions involving electrons in the same shell proceed first. If several electrons are in different shells, the Auger process involves the two electrons which are energetically closer.

(iv) Each Auger transition proceeds with the unit probability to the nearest continuum limit. The decaying electron falls to a well-established n level according to the energy equation.

(v) If the new configuration still provides a multiple excited state involving more than two electrons, those rules are applied again until only two electrons remain bound to the projectile.

(vi) If a cascading process leads to an asymmetric doubly excited state ($|n_2 - n_1| \geqslant 2$), the event is characterized as double radiative decay. Otherwise, a final Auger process takes place and the event is characterized as single charge exchange.

Finally, we note that former studies on the CTMC l distributions for different n values have shown that during the capture process the electron tries to preserve its orbital eccentricity [25]. Following this physical picture, the electronic angular momenta of the decaying electrons are determined throughout the Auger process by requiring the preservation of their respective orbital eccentricities. This condition, which in practical terms is given by $l_f = l_i(n_f/n_i)$, safely reduces the l value of the decaying electrons [initially in the (n_i,l_i) state] avoiding the risk of having unphysical l_f values greater than n_f.

III. RESULTS

In Fig. 1 we show the line emission cross sections following $Ne^{10+} + Ar$ charge-exchange collisions at an impact energy of 4 keV/amu. Such an impact energy is very close to that corresponding to the fast solar wind (~700 km/s). Present theoretical results are compared to the relative experimental data of Tawara *et al.* [22] which are normalized to our theoretical absolute values at the Ly-α peak. The data were obtained at NIST by using the EBIT trap in the "extraction mode of operation," in which the trap is used as an ion source and projectiles are accelerated into a separate collision

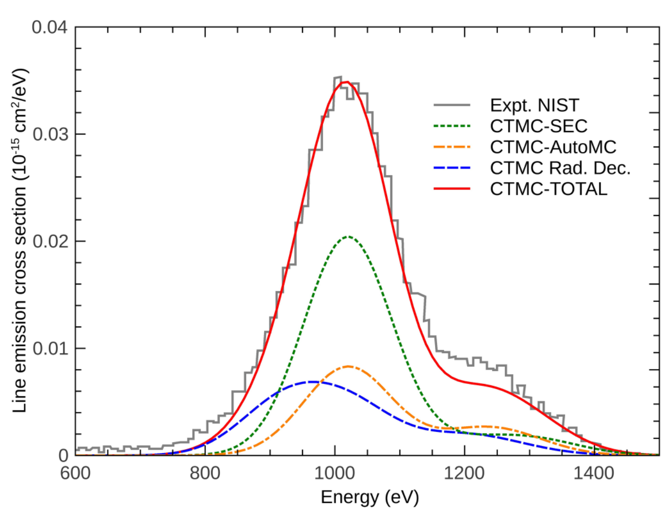

FIG. 1. (Color online) CTMC line emission cross sections for 4 keV/amu $Ne^{10+} + Ar$ collisions. Present results are compared to the NIST experimental data of Tawara *et al.* [22]. The partial contributions of true single capture SEC, autoioinizing multiple electron capture, and radiative decay channels are explicitly shown.

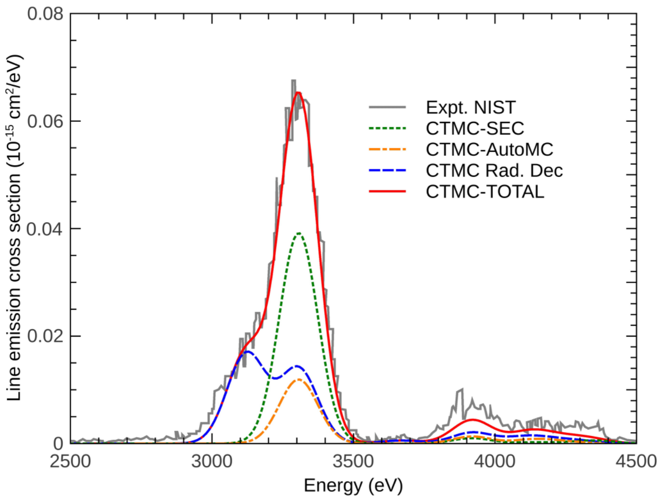

FIG. 2. (Color online) Same as Fig. 1 but for $Ar^{18+} + Ar$ collisions.

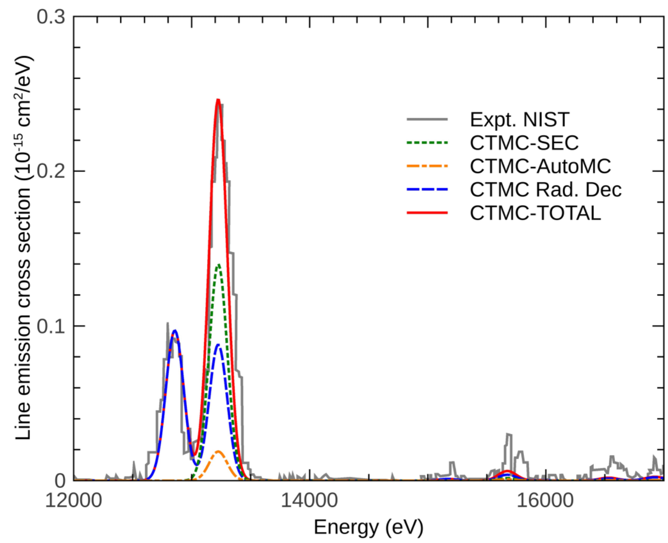

FIG. 3. (Color online) Same as Fig. 1 but for $Kr^{36+} + Ar$.

chamber. Although the reported energy resolution of the detector is roughly 130 eV for Ti Ly-α x rays at 4.5 keV, we convolute our line emission cross sections by means of 160 eV full width at half maximum (FWHM) Gaussian functions in order to match the experimental widths. The different lines show the separate contributions arising from true single electron capture with a singly charged residual target ion (SEC), and autoionizing multiple electron capture (AutoMC) and radiative decay (Rad. Dec.) channels where the target is left multiply ionized.

In this case, the theoretical fraction of x rays originating in multiple electron capture amounts up to 49.8%. This result is consistent with the experimental value of 54.1% reported by Ali *et al.* [26] using the Cold Target Recoil Ion Momentum Spectroscopy (COLTRIMS) setup at the UNReno for the same collision system at the slightly different impact energy of 4.54 keV/amu.

In Fig. 2 we now show the line emission cross sections following 4 keV/amu $Ar^{18+} + Ar$ charge-exchange collisions. In this case, the low-energy shoulder on the Ly-α peak is clearly visible. This structure, much more obvious than for bare neon projectiles, is considered a footprint of multiple electron transfer into fully stripped ions and is provided by the radiative decay channel. The doubly excited states formed in this case are stabilized through the emission of two x-ray photons, one corresponding to the hypersatellite line and one corresponding to the satellite line.

In Fig. 3 we now show the line emission cross sections following 4 keV/amu $Kr^{36+} + Ar$ charge-exchange collisions. In this case, what appeared to be a low-energy shoulder on the Ly-α peak for the Ne and Ar cases displays itself as a well-separated second peak. Here, we had to convolute our theoretical results over 180 eV FWHM Gaussian functions to match the experimental widths. We should note that the data analysis used for *in situ* EBIT measurements would not display the low-energy Ly-α peak since it would have been subtracted from the data as being due to SEC Kr^{35+} collisions.

We summarize in Table I our CTMC estimates for the relative contributions of the SEC, AutoMC, and Rad. Dec. channels to the x-ray spectra shown in Figs. 1–3. It should be noted that multiple electron capture, MEC, is responsible for a fraction which varies from 50% to 60%, which increases with projectile charge. Moreover, as the projectile charge increases, the radiative decay channel gains relevance from 25.2% for Ne^{10+} up to 54.5% of the x-ray spectrum for Kr^{36+} collisions. These results provide a clear indication on the limitations of pure three-body treatments to describe collision processes involving highly charged ions.

Now moving to lower impact energies, in Fig. 4 we consider the Berlin-EBIT data of Allen *et al.* [21] for $Ar^{18+} + Ar$ at an impact energy of 218 eV/amu. Data, in this case, were also collected using the extraction mode. The fraction of x rays originating in multiple electron capture amounts up to 51.7%. Radiative decay provides 73.2% of the CTMC MEC x rays fraction.

Figure 5 shows the lowest energy considered by Allen and collaborators in the extraction mode of operation which is 18 eV/amu. The CTMC fraction of x rays originating in MEC amounts up to 55.3%. Radiative decay provides 72.6% of the MEC fraction. This is the typical EBIT energy and x-ray spectra can also be collected *in situ* with the ions circling inside the trap. As reported in their article, these authors found strong discrepancies between the spectra obtained by using these two operational techniques.

Differences in the spectra were especially noticeable for the higher Lyman lines. Those measured *in situ* were much more

TABLE I. Relative contributions of the SEC and MEC mechanisms to the x-ray spectra shown in Figs. 1–3 following charge exchange with argon atoms at an impact energy of 4 keV/amu.

Projectile	SEC	AutoMC	Rad. dec.	MEC (AutoMC+Rad. dec.)
Ne^{10+}	0.502	0.246	0.252	0.498
Ar^{18+}	0.450	0.157	0.393	0.550
Kr^{36+}	0.399	0.056	0.545	0.601

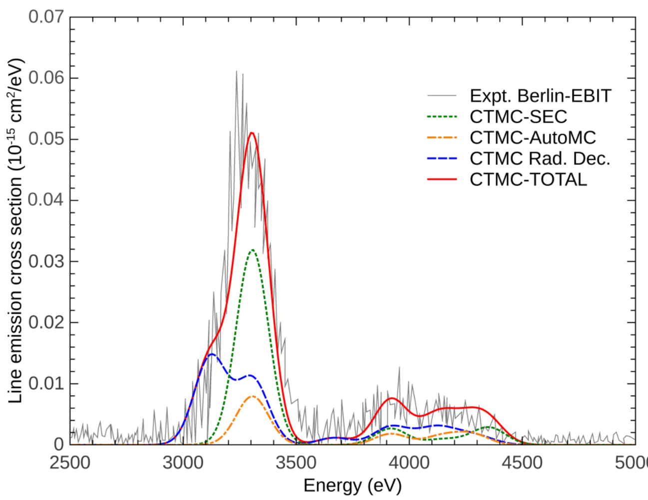

FIG. 4. (Color online) CTMC line emission cross sections for 218 eV/amu $Ar^{18+} + Ar$ collisions. Present results are compared to the Berlin-EBIT experimental data of Allen *et al.* [21]. The partial contributions of the SEC, autoionizing multiple electron capture, and radiative decay channels are explicitly shown.

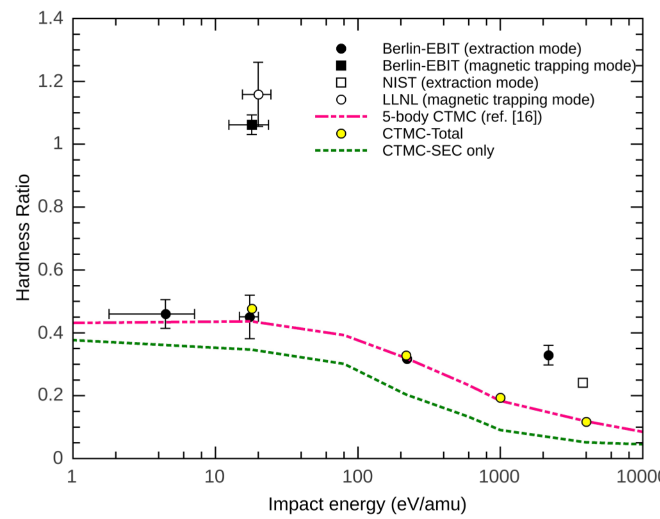

FIG. 6. (Color online) Hardness ratio as a function of impact energy for $Ar^{18+} + Ar$ collisions. Previous results obtained with a CTMC code that included three electrons [16] are shown for comparison.

prominent than those obtained in the extraction mode. This situation reflects itself strongly in the hardness ratio parameter R which is given by the ratio of x rays corresponding to the $n \geqslant 3 \rightarrow 1$ transitions to the $n = 2 \rightarrow 1$ as shown in Fig. 6. Berlin-EBIT *in situ* data at typical EBIT energies (∼20 eV/amu) are in agreement with the magnetic trapping mode results at LLNL. For the extraction mode, R values are lower by a factor greater than 2. At solar wind energies, on the other hand, the extraction mode results from the Berlin-EBIT were found in good agreement with those obtained at NIST.

Present CTMC hardness ratios are found in agreement with the extracted beam results from Berlin-EBIT. However, we note that at large impact energies our theoretical predictions tend faster to the statistical limit than the data. We also observe that the SEC channel underestimates the hardness ratio values in the whole energy range considered and the inclusion of multiple electron capture (MEC) nicely improves the agreement with the experimental extraction mode data. One thing worth noting is that SEC results should not be associated to "pure three-body results." In fact, three-body models associate to SEC contributions arising from low impact parameters that in many-electron models feed MEC channels.

Finally, we tackle the $Ar^{17+} + Ar$ collision system at an impact energy of 7 keV/amu for which there have been recent studies by Trassinelli and collaborators at GANIL [27]. Their reported relative intensities for the $1snp \rightarrow 1s^2$ transitions have been normalized to our CTMC results at the $1s3p \rightarrow 1s^2$ line. From Fig. 7 we can clearly observe that the sole contribution of the SEC channel is not enough to describe the relative intensities and that the inclusion of the AutoMC contribution drastically improves the agreement with the reported data. Radiative decay in this case does not

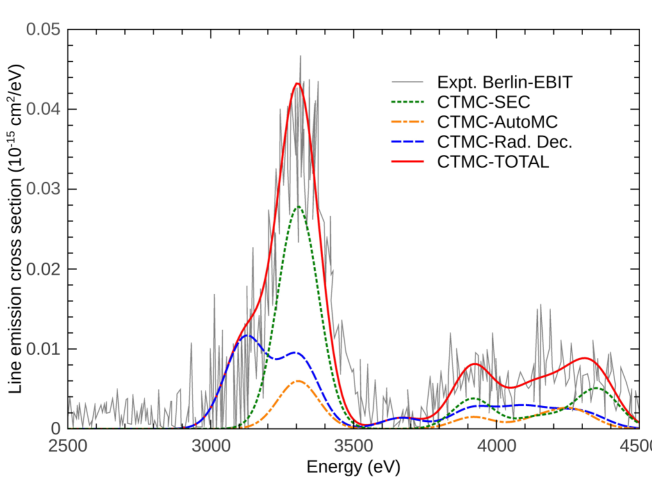

FIG. 5. (Color online) Same as Fig. 4 but for 18 eV/amu Ar^{18+} + Ar.

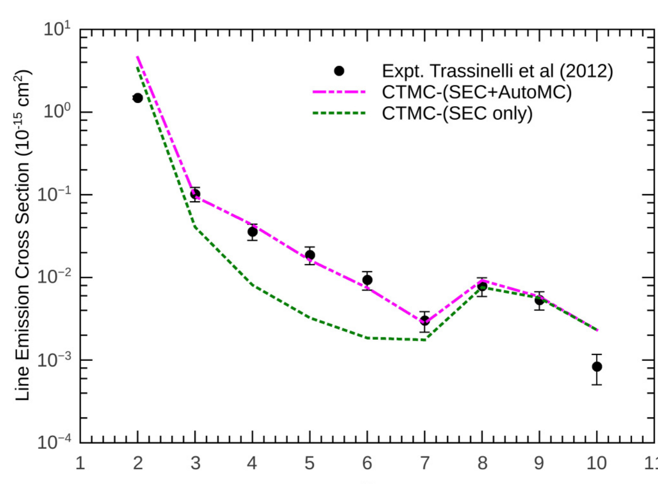

FIG. 7. (Color online) CTMC line emission cross sections for the $1snp \rightarrow 1s^2$ transitions following 7 keV/amu $Ar^{17+} + Ar$ charge-exchange collisions. The experimental data are those of Trassinelli *et al.* [27].

contribute to the present intensities since lines associated to a Li-like final projectile are well separated from those under study.

IV. CONCLUSIONS

In this work we have theoretically studied the role of the multiple electron capture channel in the x-ray spectra following charge-exchange collisions between highly charged ions and neutral argon targets. A classical trajectory Monte Carlo model based on eight noninteracting electrons sorted with sequential binding energies has been married with a decay scheme which allows the explicit separation of the multiple electron capture events in single electron capture, autoionizing multiple capture, and radiative decay. Present results for $Ar^{18+} + Ar$ at impact energies close to fast solar wind conditions are found in very good agreement with the x-ray spectra of Tawara *et al.* obtained at NIST. Our calculations nicely reproduced the low-energy shoulder in the Ly-α structure, which for larger projectile charges evolves into a well-separated peak and is due to radiative decay after double electron capture. At lower impact energies (218 and 18 eV/amu), our results were found in very good agreement with the reported data of Allen *et al.* obtained at Berlin-EBIT using the extracted beam mode of operation.

For the collision systems studied in this work, multiple electron capture is responsible for 50%–60% of the resulting x-ray spectra. Having in mind that astrophysical codes use charge-exchange (or line emission) cross sections as input data to indirectly estimate the solar wind ion abundances in remote places of the Universe, present studies suggest that the role of multiple electron capture for astrophysical gaseous targets should be revisited and/or determined.

After nearly a decade, the discrepancy among line emission spectra obtained using EBIT traps that are run in the magnetic trapping mode and the extraction mode at collision energies on the order of $\sim$20 eV/amu remains unexplained. Further joint experimental and theoretical efforts are needed in this direction to help settle this lasting and critical issue.

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